Research and Development

EPA-600/S7-81-142 Dec. 1981



Project Summary

Environmental Assessment: Source Test and Evaluation Report—Lurgi (Kosovo) Medium-Btu Gasification, Final Report

K. W. Lee, W. S. Seames, R. V. Collins, K. J. Bombaugh, and G. C. Page

An environmental data acquisition program, sponsored jointly by the U.S. EPA and the government of Yugoslavia, has been conducted in the Kosovo Region of Yugoslavia as a cooperative effort between Yugoslav and American scientists. The aim of the program was to gather data which could contribute to the characterization of potential environmental problems associated with coal gasifiers using Lurgi gasification technology. Processes involved in Lurgi gasification technology are of particular interest to the U.S. EPA because they have a significant potential for use in the U.S. The test plant, though not representative of state-of-the-art pollution control practice, provided process discharge streams that are typical for Lurgi gasification technology.

An extensive sampling and analysis program was conducted at Kosovo. The main thrust of the study was to characterize the discharges of the plant's key processes and to prioritize pollutant discharges in terms of their potential for causing adverse health and/or environmental effects. Prioritization was accomplished using the Source Analysis Model/IA (SAM/IA) developed by EPA's Industrial Environmental Research Laboratory (IERL)

at RTP. The prioritized data provides a basis for making engineering judgments on control technology.

The major conclusions drawn from this study are that, without environmental controls, a Lurgi type process exhibits a significant potential for polluting the environment; that virtually all discharge streams, whether gaseous, aqueous, or solid, present a significant potential for transferring pollutants from the process to the environment: and that particulates in gaseous discharges are carriers of potentially hazardous PNAs. Based on the SAM/ IA prioritization, the Kosovo discharge streams of highest concern are: H2Srich waste gas, extracted phenolic water, and heavy tar. Gasifier ash was found to be of low concern. Discharge stream pollutants of major concern are sulfur species and aromatic hydrocarbons, including polycyclic aromatics.

Trace elements were found to be of much lower significance than trace organics.

This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

The purpose of this program was to characterize the discharge streams from a commercial Lurgi-type coal gasification facility as a means of determining the pollution potential of Lurgi type processes. The program was designed in response to a need for representative data on potential environmental problems associated with the commercial application of Lurgi coal gasification technology. The Lurgi coal gasification process was selected for study because it has a significant potential for use in the U.S. and is being considered as a commercially feasible technology for the production of substitute natural gas and for the indirect liquefaction of coal. The Lurgi type plant chosen for this study is in the Kosovo region of southern Yugoslavia. It uses 3.5 m diameter gasifiers. The opportunity to make a comprehensive environmental characterization of an operational, commercial-scale, Lurgi type coal gasification plant was considered valuable since a number of U.S. companies have announced plans to construct such plants. Thus, characterization of selected process and discharge streams for the Kosovo plant provided a valid insight into problems that must be considered by U.S. designers in developing process modifications and/or control schemes necessary to meet U.S. environmental standards.

The program, conducted over 3 years, was a joint effort among scientists from the U.S. and Yugoslavia. The participating organizations, and their roles, are shown in Table 1.

The program consisted of four phases:

Phase	Objective
1	Identify and measure major and minor pollutants in discharge streams.
2	Identify and measure trace pollutants in discharge streams.
3	Characterize ambient air pol- lutants in the vicinity of the

4 Measure fugitive emission rates from leak sources in the plant (cofunded by DOE).

nlant

This report presents information on major, minor, and trace pollutants in the discharges of the Kosovo Lurgi type plant. Results from testing Phases 1 and 2 were combined to provide a "best value" for use in evaluating the discharges. This report includes an assessment of specific discharges and also an evaluation of discharge severity as determined by EPA/IERL's SAM/IA model for prioritizing pollutants on the basis of their estimated potential for causing adverse health and ecological effects.

Plant Description

The Kosovo gasification plant is a part of a large mine-mouth industrial complex that includes a coal mine, a coal preparation plant, a coal gasification plant; an air separation plant, an ammonia plant, a power generating plant, and a steam generating plant. The gasification plant consists of six operational units as illustrated in Figure 1. The plant consumes dried lignite and

produces two primary products (a medium-Btu fuel gas and hydrogen) and four by-products (tar, medium oil, naphtha, and crude phenol). The plant's operation is as follows: run of the mine coal, brought from a nearby open-pit mine, is dried by the Fleissner process and sized to select particles 6 - 60 mm in diameter. The dried coal is fed to the Lurgi type gasifiers where it is reacted with steam at 2.5 MPa (25 atm) pressure to produce a crude gas which is quenched, cooled, and then cleaned by the Rectisol process before being transported to the utilization site. As the crude gas is guenched and cooled, tars, oils, and phenols are removed in a stream with the gas liquor (phenolic water). In the gas-cleaning operation, condensible organics are removed by refrigeration, after which the acidgases (H2S and CO2) are removed by sorption in cold methanol. The acid-gasrich methanol is regenerated by depressurization and heating, releasing a gas rich in H2S (which is flared) and a gas rich in CO2 (which is vented to the atmosphere).

Tars and oil are separated from the phenolic water by decantation after which the water soluble organics (crude phenols) are extracted with diisopropyl ether. Ammonia, removed from the phenolic water by steam-stripping, is vented to the atmosphere.

The Kosovo gasification plant is a commercial-scale facility. Figure 2 shows the design flow rates of the plant's major inlet and outlet streams. These data indicate that the plant is designed to produce 25 Mg (65,000 m³ at 25°C) of product gas for every 80 Mg of dried coal consumed. Although this plant is smaller than proposed firstgeneration U.S. Lurgi gasification facilities, it contains many of the process units which are likely to be employed in future U.S. Lurgi plants. For this reason, the plant is felt to be representative of many aspects of the Lurgi gasification facilities which are being considered for commercialization in the U.S. However, the environmental control practices followed at the Kosovo plant are not representative of proposed U.S. Lurgi plant designs. Many of the plant's waste streams are controlled but none of the controls would be characterized as best available by current U.S. standards. Thus, while the discharges that enter the environment at Kosovo are not representative of those that would be encountered in similar U.S. facilities, the types of control problems

Table 1. Functions of Various Organizations Participating in the Kosovo Test Program

Organization	Location	Function
EPA/IERL	Research Triangle Park North Carolina	Funding Agency, Project Director
Radian Corporation	Austin, Texas	Prime Contractor and Coordinator
Rudarski Institut	Belgrade, Yugoslavia	Sampling/Analyses/Data Analyses/Overseas Coor- dinator
Kombinat Kosovo	Obilic, Yugoslavia	Plant Operation/Sampling
Kosovo Institut	Obilic, Yugoslavia	Sampling/Trace Element Analyses
Institut za Primenu Nuklearne Egergije	Belgrade, Yugoslavia	On-Site GC Analyses/ Organic Analyses

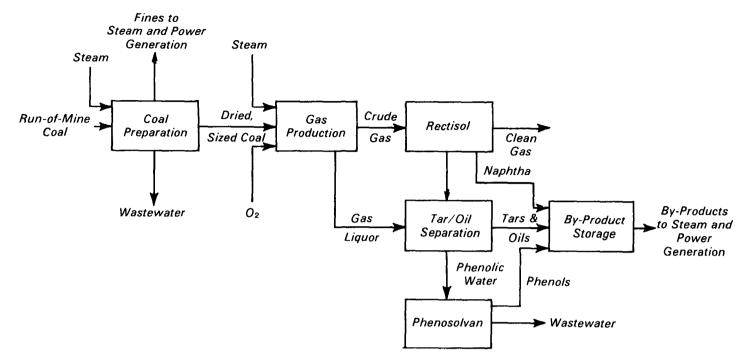


Figure 1. Simplified flow diagram of the Kosovo coal preparation/gasification plant operations.

facing U.S. Lurgi plant operators will be similar to those found at Kosovo. A study of the waste streams data generated at Kosovo, therefore, should aid U.S. plant designers in developing the process modification and control schemes necessary to achieve U.S. control standards.

Stream Selection and Test Methods

The Kosovo test program was conducted over 3 years as a phased effort to characterize the plant's major, minor, and trace pollutant discharges, Initially, screening studies were conducted to select the most significant streams for more detailed characterization, Stream selection was based on: high discharge rate, high pollutant concentrations. trace pollutants of high concern, and important process information. Streams exhibiting a high discharge rate were selected for study because at high flow rates, even very low concentrations of moderately toxic pollutants could result in a significant environmental burden.

Streams exhibiting a moderate or low discharge rate were selected for study if they contained high pollutant concentrations of pollutants of high concern.

Some streams were selected to provide information on the fate of trace elements and trace organics throughout

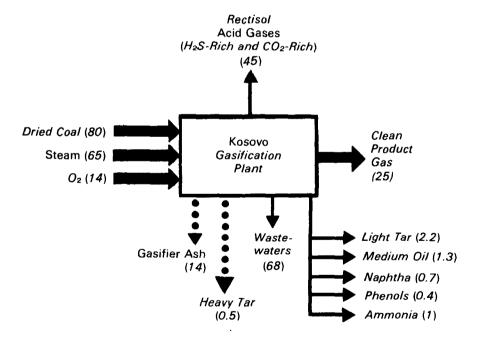


Figure 2. Design flow rates of key streams in the Kosovo gasification plant (all values in Mg/hr based on five gasifiers in service),

the process; and some streams were selected to provide a better understanding of process operations that affect the distribution of pollutants in those processes.

The streams considered in the program are shown in Table 2; of these streams.

50 were tested (35 gaseous, 5 aqueous, 6 solid phase, and 4 organic liquid) and 31 (17 gaseous, 5 aqueous, 6 solid phase, and 3 organic liquid) received detailed characterization.

The detailed characterization program addressed the following parameters:

Gaseous Streams

- Flow rate.
- Particulate concentration.
- Gas composition.
- Condensible organics.
- Trace elements content.

Aqueous Streams

- Water quality parameters.
- Trace elements content.
- Organic constituents.

Solids

- Proximate analysis.
- Ultimate analysis.
- Trace elements content.
- Leachate composition.

Liquid By-Products

- Bulk composition.
- Trace elements content.
- Polynuclear aromatics content.

Methods used to study these parameters are summarized in Tables 3 through 7.

Bioassays were performed on samples from seven streams: dry gasifier ash, heavy tar, light tar, medium oil, naphtha, Phenosolvan inlet water, and Phenosolvan wastewater. These tests provided information about potential biological effects from the Kosovo plant's solid, liquid by-product, and wastewater streams.

Sampling Methods

Sampling methods used in this study are listed in Tables 3 through 7. Gas stream sampling was the most complex and consumed an estimated 90 percent of the sampling effort. Although published methods were followed in principle, a considerable amount of adaptation was needed to fit the collection requirements. Problems encountered in sampling included: high stream pressure, widely pulsating stream flow rates, entrained mist, high levels of condensable organics, and reactive sample components.

Particulate sampling was considered the most difficult because the entrained mist and condensables present in many streams obstructed the filter in a conventional collection train. For such streams, particulates were collected in a train of water-filled impingers. The collected matter was isolated from the impinger liquid (by a combination of extraction, filtration, and evaporation) and determined gravimetrically as tars and oils, filterable solids, and dissolved solids.

Table 2. Number and Type of Significant Streams in Each Section of the Kosovo Plant

		pe or Stream	n		
Plant Section	Gaseous	Aqueous	Solid	Organic Liquid	Total
Coal Preparation	3	1	2	0	6
Gas Production	8	1	3	0	12
Rectisol	5	1	0	1	7
Tar/Oil Separation	7	1	1	2	11
Phenosolvan	12	3	0	2	17
By-Product Storage	<u>6</u>	<u>o</u>	0	<u>o</u>	<u>_6</u>
	41	7	6	5	<i>59</i>

Table 3. Measurement Methods for Flow Rate

Parameter	Measurement Method	Analytical Method
Velocity	Pitot	_
Temperature	Thermocouple	_
Pressure	Manometer/Gauge	_
Molecular Weight	Gas Collection & Conditioning system	TC-GC

Table 4. Measurement Methods for Particulates

Component	Collection Method	Analytical Method
Suspended Particulates	Filtration - in stack at duct temperature	Gravimetric
	out of stack at 250°F (121°C)	
Suspended Particulates & Condensables (tar-laden streams)	Cold Impinger	Filtration/Extrac- tion/Gravimetric

Table 5. Measurement Methods for Gas Composition

Component	Collection Method	Analytical Method
Moisture	Impinger/Ice bath	Gravimetric
Fixed Gases*	Gas Collection and Conditioning System/ Glass Bombs	TC-GC
Hydrocarbons**	Same as Fixed Gases	FID-GC
Sulfur Species***	Same as Fixed Gases	FPD-GC
H₂S	Impinger/CdOAc	Titration
HCN	Impinger/NaOH	Distillation/Titration
NH ₃	Impinger/H₂SO₄	Distillation/Titration
Phenols	Impinger/NaOH	Colorimetric

 $^{^*}CO$, CO_2 , O_2 , H_2 , CH_4 , N_2 $^{**}C_1$ - C_6 , C_6 , BTX $^{***}H_2S$, COS, RSH, SO_2

Table 6. Measurement Methods for Condensable Organics

Component	Collection Method	Analytical Method
Condensable Hydrocarbons (xylene and up)	Condensation/XAD-2 Resin	Extraction: TCO GRAV IR
		LC Fractionation: TCO GRAV IR GC-MS
Benzene, Toluene, and Xylene	Charcoal Tube	Extraction/FID-GC

Table 7. Measurement Methods for Trace Elements

Group	Collection Method	Analytical Method
Nonvolatile	Acid/Base Impinger	Dissolution/AA,
Elements*	Series	Graphite Furnace
Volatile	Acid/Base Impinger	Dissolution/AA,
Elements**	Series	Hydride Generation
Carbonyls***	Filtration/HCI Impingers	AA, Graphite Furnace

^{*}Be, Cd, Ce, Cr, Cu, Mo, Ni, Pb, Sr, Tl, V, Zn

Trace elements, volatile phenols, and reactive species (including NH₃, H₂S, and HCN) were also collected from gas streams by impinger train. Gas samples for gas chromatographic analysis were collected in 0.5-L silanized glass bombs at 0.5 atm pressure with the aid of a specially built apparatus that included a heated inlet, a permeation drier, and a diaphragm pump.

Analysis Methods

Analysis methods used in the study are also listed in Tables 3 through 7. All gas analyses were performed on-site. Fixed gas analyses were run on all GC samples to detect air leakage and to obtain the compositional information provided by the analyses. Any sample showing an abnormal level of air was discarded and the sampling repeated until a valid sample was obtained from the collection point.

Test Results

Composition data were obtained on 49 streams which included discharge, feed, product, and process streams. Detailed composition and flow rate data were obtained in 19 gaseous, 2 aqueous, and 2 solid discharge streams. Results were used to rank both discharge streams and pollutants in terms of their potential for causing adverse health or

ecological effects according to the EPA-IERL SAM/IA model. Of the three discharge media, gas streams were found to be the major carrier of pollutants in terms of both pollutant mass and discharge severity (DS). Discharge severity was computed in terms of both DS_H (potential for adverse health effects) and DS_E (potential for adverse ecological effects).

Gaseous Discharge

The most significant gaseous discharge stream in the Kosovo gasification plant, in terms of DSH, is the H2S-rich waste gas from the two-stage Rectisol plant (at Kosovo, this stream is flared). The next most significant stream is the ammonia stripper vent discharge which contains 42 percent ammonia and high concentrations of H2S, phenols, HCN. and mercaptans (this stream has the highest DS_E). The CO₂-rich waste gas, which is the largest atmospheric discharge stream at Kosovo, releases significant quantities of hydrocarbons and reduced sulfur species. Although the DS of this stream is 2.5 percent of the H₂S-rich stream, it is environmentally very significant because of its high flow rate.

Tank vents were found to be significant sources of pollution. Although their flow rates were small (<5 m³/hr), their

pollutant concentrations were high (up to 4 percent benzene).

The major pollutant class, based on discharge mass, was light hydrocarbons. The most significant class in terms of Weighted Discharge Severity was sulfur species.

Particulates in Gaseous Discharges

Particulate loadings in the discharge from both the start-up vent and the low pressure (LP) coal lock vent were about 10 times higher than in the discharges from the other vents that were tested. About 90 percent of the particulate loading in these discharges consisted of condensable organics (tar and oils) which contained appreciable amounts of PNAS. It is estimated that 500-1500 μ g/m³ benzo(a)pyrene is contained in the LP coal lock discharge, resulting in an estimated discharge rate of 3-9 μ g/sec.

Aqueous Discharge

Aqueous discharges from both Phenosolvan and gasification had high values for solids, COD, and permanganate value. Gasification wastewater exceeded effluent guidelines for suspended solids by 87/1. Phenosolvan effluent, the plant's largest waste stream, exceeds effluent guidelines in both Total Suspended Solids and pH. The major pollutant of this discharge was phenol. Although the Phenosolvan process removed 90 percent of the incoming phenol, it removed only 70 percent of the total organic carbon.

Solid Discharges—

Gasifier ash, the plant's major solid waste, presents no unusual environmental problems. Although it has a positive heating value, it would not be considered ignitable by RCRA standards. Trace element levels in ash leachate were below the concentration levels that would cause gasifier ash to be considered toxic according to RCRA limits.

Heavy tar, which has a higher heating value than the feed coal, contains polycyclic aromatic hydrocarbons including a significant concentration of benzo(a)pyrene (0.024 percent).

Comparison of All Discharge Media—

A comparison of 18 discharge streams in all media (gas, aqueous, and solid), shown in Figure 3, indicates that the

^{**}Hg, As, Sb, Se

^{***}Ni. Fe

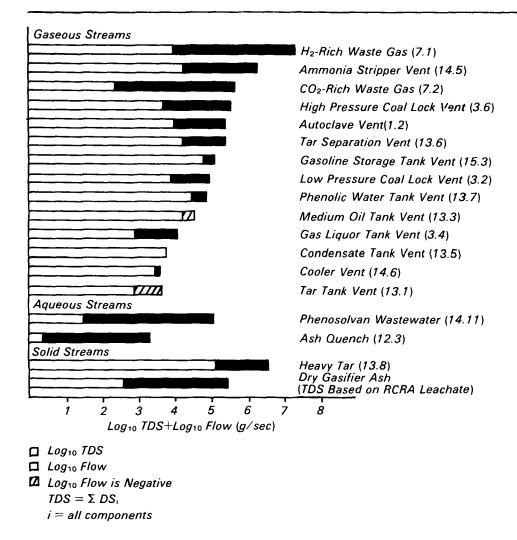


Figure 3. A comparison of the total weighted discharge severity values (health) key Kosovo gaseous, aqueous, and solid streams.

streams of highest environmental concern, in terms of health, are the H₂S-rich waste gas, the heavy tar, and the ammonia stripper vent discharge, respectively.

Bioassay Results-

Of the nine samples tested, the liquid by-products were the most toxic in rodent toxicity tests. By-product tar and phenolic water each gave a positive response to the Ames test for carcinogenicity. However, the Phenosolvan effluent water was negative, indicating a reduction in bioactivity by the process. Gasifier ash and ash leachate showed little or no biological activity.

Mass Balances—

Mass balance calculations on the test data show that most of the major elements in the feed coal are discharged

in the gaseous streams. The major transporter of sulfur is the H_2S -rich waste gas stream; of nitrogen, the ammonia stripper vent; and of carbon, the product gas and a combination of the H_2S - and CO_2 -rich waste gas streams. Accountability of the sulfur, nitrogen, and carbon was 180, 51, and 92 percent, respectively.

K. Lee, W. Seames, R. Collins, K. Bombaugh, and G. Page are with Radian Corporation, P.O. Box 9948, Austin, TX 78766
William J. Rhodes is the EPA Project Officer (see below).

The complete report, entitled "Environmental Assessment: Source Test and Evaluation Report—Lurgi (Kosovo) Medium-Btu Gasification, Final Report," (Order No. PB 82-114 075; Cost: \$24.00, subject to change) will be available only from:

National Technical Information Service 5285 Port Royal Road Springfield, VA 22161 Telephone: 703-487-4650

The EPA Project Officer can be contacted at: Industrial Environmental Research Laboratory U.S. Environmental Protection Agency Research Triangle Park, NC 27711

United States Environmental Protection Agency Center for Environmental Research Information Cincinnati OH 45268 Postage and Fees Paid Environmental Protection Agency EPA 335



Official Business Penalty for Private Use \$300

> PS 0000329 U S ENVIR PROTECTION AGENCY REGION 5 LIBRARY 230 S DEARBORN STREET CHICAGU IL 60604